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A MODIFIED STOKESIAN DYNAMICS METHOD FOR MINERAL SUSPENSIONS

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Abstract. A 3-dimensional modified Stokesian dynamics-based technique for simulating mineral particle suspensions is presented. Stokesian dynamics is a mesh free particle approach, which resembles the discrete element method. It includes hydrodynamic interactions and other interparticle forces.

Expressions for the hydrodynamic interactions were modified based on results from finite element (FE) calculation. The modifications allow for broader particle size distributions than captured by traditional analytical expressions describing hydrodynamic interactions. In addition, models are presented for colloidal interactions, steric repulsion caused by polymer adsorbed onto mineral particles and the Brownian motion. These models expand the applicability down to μm and nm size particles. Comparison between governing forces can be made by generation of dimensionless expressions such as the particle Reynolds and Peclét numbers.

Numerical simulations performed using this technique enable the study of microscopic scale mechanisms and the characterisation of particle systems. This allows for appreciation of microstructure development in time and the prediction of macroscopic level properties of particle suspensions and consolidating systems.

This paper reports on both model development and results utilising the above-described approach. In conclusion, the method is put into context by discussion of the applicability of the method in various wet-state mineral processing applications.

1 INTRODUCTION

Particulate systems are of relevance in many industrial processes and products. To name but a few, mineral processing, paper manufacturing, printing, paint production and many biological and pharmaceutical applications are based on processing and handling of particulate matter. Products can be in various forms, such as pellets, briquettes, powders or

tablets. Furthermore, during processing or as an end product, the material may exist in the form of a liquid suspension. This is exemplified by paint, paper coatings and printing inks.

Empirical studies of particle-based processes can in many cases be difficult or sometimes virtually impossible. This can be due to small size and time scales, intense external conditions (forces, pressures, temperatures, velocities) and complex particle interactions and system geometries. The understanding of liquid suspensions requires knowledge on both physical and chemical phenomena. It is often not possible to distinguish one effect from another, and therefore difficult to experimentally evaluate the relative influence of phenomena involved. In this respect, one can resort to numerical simulation for increasing ones understanding of the behaviour of materials in such processes. Simulation allows for adjustment of individual parameters and the possibility to explore the separate or combined influence of competing interaction models.

In mineral processing, particle simulation has traditionally been geared towards studies of dry-state granular material behaviour in complex geometries. Some examples include storage in silos, hopper discharge and conveyor transport processes [1]. Interparticle mechanical interactions and response to gravity and equipment behaviour (e.g. belt feeder translation) has in these cases often been sufficient in describing processes with relative accuracy. In more recent research, however, the focus has turned towards comminution and separation processes. In many cases, the models used for material transport simulation were simply transferred to the new fields of interest. To an extent it has been possible to gain improved understanding by this approach. There is a need, however, to improve models in terms of material property definitions and inclusion of more complex interactions and phenomena. There exist several examples of applications where such aspects which are now gaining relevance in mineral processing have been much more extensively studied. It is deemed possible to benefit from these interdisciplinary differences for improving simulation methodologies in mineral industry applications.

In a suspension, the behaviour of particles is governed by the macroscopic flow of liquid phase, hydrodynamic interparticle, particle-solid boundary and particle-free surface interactions. Colloidal interactions, the Brownian motion and various forces induced by the presence of polymers or other additives, are of relevance especially for nm to µm size particles. External field forces, as resulting from e.g. gravity and magnetic susceptibility of particles, can also play a significant role for the properties and behaviour of the suspension.

In this paper, the Stokesian dynamics method is introduced as an approach for simulating the behaviour of particles in suspension. Modifications to the original Stokesian dynamics approach are discussed. This includes adjustment of the hydrodynamic expressions in order to improve calculation accuracy for systems of broad particle size distribution and high solids concentration. Furthermore, models for various types of non-hydrodynamic interactions are presented. In a case study-type approach, the results of some studies utilising the models are discussed. Finally, areas of mineral processing are identified, where there could be benefits of using the Stokesian dynamics methods for increasing the understanding of particle-level phenomena and processes.

2 METHODOLOGY

Stokesian Dynamics is similar to Brownian Dynamics and the Discrete Element Method. DEM is typically applied on systems of a larger size scale than SD, which is strongly leaned towards liquid suspensions and colloidal-size interactions [2]. Colloidal interactions are of importance for nm to µm size particles in suspension. One major difference is that Stokesian dynamics includes expressions for taking into account various types of hydrodynamic interactions [3]. Furthermore, some type of coupling between the particles and liquid phase is needed to simulate flow in complex geometries. One approach is to import CFD/ FEM-generated flows to SD, and therefore obtain realistic liquid velocities and profiles [4]. Although the suspending medium is typically assumed to be a Newtonian liquid, the combination of particles and liquid gives the suspension viscoelastic properties. The rheology will then be determined by the particle interaction models and parameters used. As indicated above, the method does not typically include back-coupling between the solid and liquid phases. Thus, macroscopic flow fields and interparticle liquid flow influence particles by forces and torques which induce translation and rotation on particles, but the particles themselves do not influence the flow of liquid. Full coupling can be obtained, but comes with added complexity and increased computational expense.

Stokesian dynamics is based on the N-body Langevin equation,

$$\mathbf{m}\frac{d\mathbf{U}(t)}{dt} = \mathbf{F}^{H} + \mathbf{F}^{P} + \mathbf{F}^{B}, \tag{1}$$

which is simply a variant of Newtons second law of motion. It states that the sum of all forces acting on a particle must balance with its mass and acceleration. The above forces are of different types. $\mathbf{F}^{H}$ carries information on the hydrodynamic forces. These forces depend not only on particle positions but also on their velocities relative to each other and any externally applied flow field. $\mathbf{F}^{P}$ are interparticle forces (e.g. colloidal, steric and other forces) which generally depend on interparticle distances and $\mathbf{F}^{B}$ are stochastic single-particle forces e.g. as result of thermal vibration. If the particle Reynolds number is small, which is the case for small particles with low velocity difference relative to the continuous phase, an additional assumption can be made. In this event, the inertia of particles can be considered insignificant and the left side of equation (1) can be put as zero. This implies that the response of particles to the flow of surrounding liquid is instantaneous and that particles accelerate immediately to balance the force equation. Due to the complexity of interactions, SD-particles have with few exceptions been regarded as rigid spherical particles. There are examples, however, where ellipsoidal particles have been used. Ellipsoids are one of few cases of non-spherical particle shapes where exact mathematical expressions can be obtained for numerical calculation [5].

Below, models for hydrodynamic, colloidal and steric interactions are discussed. The effect of Brownian motion is included as a stochastic force. Additional interactions resulting from the presence of polymer in particle suspensions (bridging and depletion flocculation) are not considered in this work.

2.1 Hydrodynamic forces

In this chapter, the hydrodynamic forces are only briefly discussed. Further details on the hydrodynamic interactions have been presented by Nopola and Sand et al. [4,6].

Particle movement in a suspension is typically driven by the macroscopic flow of the liquid phase, often referred to as Stokes drag. Additional hydrodynamic interactions result from the movement of particles relative to each other, solid boundaries or to the free surface. One can separate between four different modes of particle relative motion; pumping, shearing, twisting and squeezing motion. A visualisation from the numerical simulations is shown in Figure 1.

The total net hydrodynamic force and torque on particles as result of the relative motions are calculated between each particle and nearby particles as given by the neighbour list.

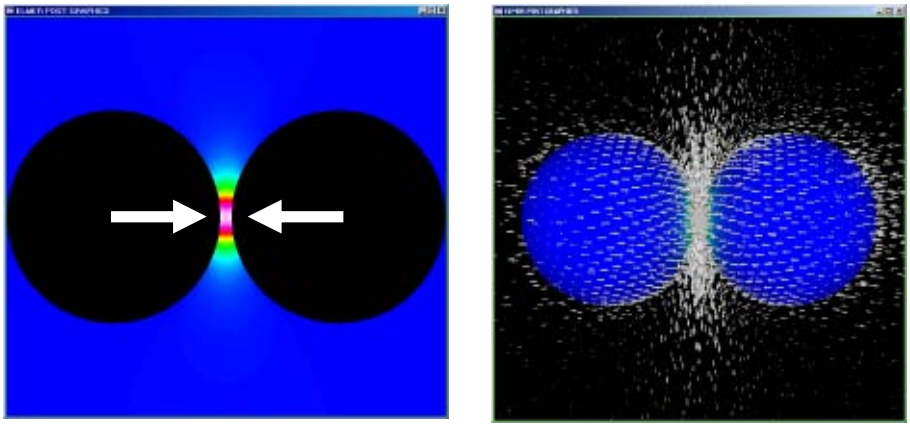

Figure 1. Squeezing relative motion as simulated and visualised using Elmer. Pressure (left) and liquid velocity vectors (right) [6].

Analytical expressions for the modes of relative motion has been presented by Kim and Karrila [5]. These expressions, however, are only suitable for quite narrow particle size distributions. For particle size ratios beyond $\beta = 0.1$, an alternative approach needs to be used. Therefore, numerical simulations were performed based on finite element analysis and the results could be compared with the analytical expressions. The theoretical equations were then fitted with the numerical results in order to find better approximations of the forces and torques. A comparison between numerical results, analytical results by Kim and Karrila and modified expressions for the shearing relative motion is shown in Figure 2 [4-6].

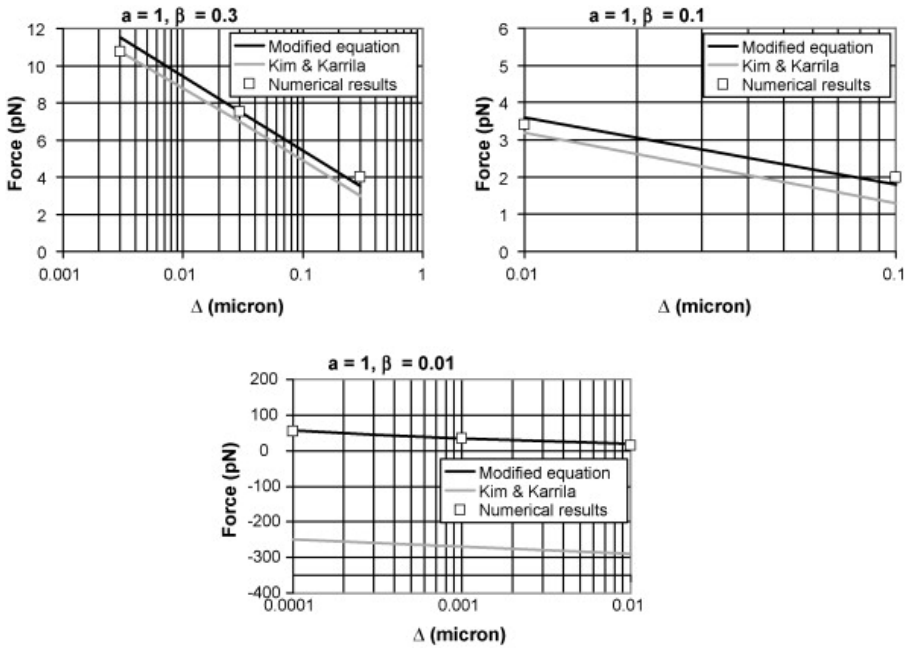

Figure 2. Comparison of numerical Kim and Karrila analytical expressions and modified expressions for the shearing relative motion [6].

In most cases good agreement between the analytical and numerical solutions was found. However, especially for large particle size differences and small interparticle surface separation distances, significant differences could be observed.

Details on this procedure as well as the fitted equations are listed in Nopola and Sand et al. [4,6]. Furthermore, particle/ solid boundary och particle/ free surface interactions are described by Sand et al. [6].

2.2 Non-hydrodynamic forces

Colloidal interactions

The colloidal force between particles is a result of the conformation and distrubution of ions and molecules of the suspending medium in response to the surface charge of particles. The colloidal interaction between two particles in a suspension has been formulated by the DLVO-theory (Derjaguin, Landay, Verwey. Overbeek) and is typically composed of an electrostatic repulsion and a van der Waals attraction component. The force between two charged particles can therefore be attractive or repulsive depending on the colloidal interaction parameters and the interparticle distance. The net colloidal interaction force is expressed as the sum of the electrostatic and van der Waals components

$$F_{coll}^{P} = F_{el}^{P} + F_{vdw}^{P}. \tag{2}$$

The electrostatic repulsive component is given by

$$F_{el}^{P} = 4\pi\kappa\varepsilon_r\varepsilon_0\psi_1\psi_2\frac{a_1a_2}{a_1+a_2}\frac{e^{-\kappa\Delta}}{1+e^{-\kappa\Delta}}, \tag{3}$$

where κ is the reciprocal double layer thickness, ε_r the dielectric constant of the continuous phase, ε_0 the permittivity of vacuum, ψ_1 and ψ_2 the surface potentials of the interacting particles and Δ the surface separation distance. a_1 and a_2 are the radii of the interacting particles [7]. The van der Waals attractive force can be described by

$$F_{vdw}^{P} = -A_H\frac{\beta}{1+\beta}\frac{\lambda(\lambda+22.232\Delta)}{6\Delta^2(\lambda+11.116\Delta)^2}, \tag{4}$$

where A_H is the Hamaker constant, λ the London characteristic wavelength and β the size ratio between the larger and smaller particle [7].

Steric repulsion

Polymer dispersants are often added as a stabiliser to mineral suspensions. As polymer is adsorbed onto mineral particles they create a steric barrier which helps to increase interparticle distance and prevents agglomeration. The resulting steric force between particles is modelled as an osmotic pressure difference for surface separation distances less than twice the thickness of the adsorbed polymer layer. If the distance is less than the thickness of one polymer layer, an additional elastic force comes into play. This additional force is assumed to result from the compression of polymer chains, as the steric boundary layers overlap.

Thus, the steric force is described as

$$F_{st}^{P} = F_{osm}^{st} + F_{el}^{st}. \tag{5}$$

The osmotic, F_{osm}^{st}, and elastic, F_{el}^{st}, components are calculated as

$$F_{osm}^{st} = \begin{cases} 0 & \Delta \geq 2\delta_{ster} \\ K_1a(\Delta - 2\delta_{ster}) & \delta_{ster} < \Delta < 2\delta_{ster} \\ K_1a\left(\delta_{ster} - \dfrac{2\delta_{ster}^2}{\Delta}\right) & \Delta \leq \delta_{ster} \end{cases} \tag{6}$$

$$F_{el}^{st} = \begin{cases} 0 & \Delta \geq \delta_{ster} \\ K_2a\delta_{ster}\ln\left(\dfrac{4\delta_{ster}^3}{\Delta(\Delta-3\delta_{ster})^2}\right) & \Delta < \delta_{ster} \end{cases} \tag{7}$$

where Δ is the surface separation, δ_{ster} the polymer layer thickness. K_1 and K_2 are constants that depend on the effective volume fraction of polymer in the adsorbed layer and density,

molecular weight and solvency of the adsorbed polymer, and the other parameters as defined earlier [7].

Brownian motion

Brownian motion results from thermal vibration of particles and collisions with molecules of the suspending medium. The Brownian motion model is based on the Einstein equation for calculation of the mean Brownian displacement, $\bar{x}$, of a particle as function of time, t, as

$$\bar{x} = \sqrt{2Dt} \,. \tag{8}$$

Particle diffusivity, D, is calculated as

$$D = \frac{RT}{6\pi\mu a N_A}, \tag{9}$$

where T is the system temperature, μ the viscosity of the continuous phase, R the universal gas constant and N_A Avogadro's constant [7].

In this work, Brownian motion is produced by applying a force with random direction to each particle at every time step. The magnitude of the force is calculated with the help of a iteratively determined fitting parameter that takes into account system temperature and particle size range. The model showed good compliance with analytical results. More details on the model can be found in [6,8].

2.3 Analysis of Governing Forces

The interactions and forces described above can be evaluated in order to determine which models are relevant to include in simulations. This is done by forming ratios between forces, which will indicate which forces dominate over others. Values typical for the particle system and process under study should be used as parameters. This type of comparison can be very useful in understanding which models have the most influence on the process under investigation. Conversely, one can obtain indications whether some forces are insignificant and possible to exclude from the calculation. Common dimensionless numbers include the particle Reynolds and Peclét numbers [8].

The impact of hydrodynamic forces of the suspending medium on the particles can be described using the particle Reynolds number, Re_p. The particle Reynolds number can be calculated using the equation

$$\mathrm{Re}_p = \frac{\rho_l a_{char} u_{char}}{\mu}, \tag{10}$$

where ρ_l is the density of the continuous (liquid) phase, a_{char} the characteristic particle size, u_{char} the characteristic velocity of the particles relative to the liquid and μ the viscosity of the suspension (Brady et al. 1988). In Stokesian Dynamics, the particle Reynolds number plays an important role due to the lubrication approximation, which assumes particles to react instantaneously to the flow of liquid. This approximation is valid for small particle Reynolds numbers.

The Peclét number, Pe, is used to compare the convective effect with the effect of particle diffusion. Convection is given by flow of the continuous phase, while diffusion results from Brownian motion. The Peclet number can be expressed as

$$Pe = \frac{6\pi\mu a_{char} Hu_{char}}{kT} \quad , \tag{11}$$

where H is the characteristic length scale, k the Boltzmann constant and T temperature. The other parameters are as defined earlier. The characteristic length scale is chosen such, that the movement of particles over this length scale would have significant effect on the microstructure or properties of the particle system.

A similar dimensionless expression can be established for comparing the relative influence of hydrodynamics in relation to colloidal interactions. The expression, here termed Kc, can be written as

$$Kc = \frac{a_{char}\mu u_{char}}{F_{el}^{P} + F_{vdw}^{P}} , \tag{12}$$

where, F_{el}^{P} is the electrostatic repulsion and F_{vdw}^{P} the van der Waals attraction component of the DLVO interaction model.

What furthers complicate the use of these types of ratios is that conditions often change in dynamic simulation, both with geometry, particle size distribution and time. In such cases it is useful to establish intervals, to determine how the influence of various forces change with time or system properties.

3 SIMULATION RESULTS

In the following section we shortly review some studies making use of the modified Stokesian dynamics method, as described in this paper. In all cases, the properties of the particle systems are set to correspond to fine-grade mineral suspensions.

Filter cake properties

The structure of filter cakes as function of particle size, dewatering rate and temperature was studied [8]. The particle size range was 0.2 to 2 μm and a polydisperse system was also the subject of investigation. One can anticipate the filter cake structure to arise due to the ability or lack of ability of particles of each size and density fraction, to migrate against an absorption flow of liquid phase through a filter boundary. Assuming that only liquid is allowed to penetrate through the base substrate, the temperature and particle size-dependent Brownian motion will compete against the flow rate of the liquid. The solids profile in response to liquid flow rate, particle size, and temperature could be reported. Furthermore, size segregation effects could be studied for a polydisperse size distribution.

The mechanism of solids structure formation in the filtering process that takes place during paper coating consolidation has been a subject of dispute in the literature. One of the main results of this investigation was to show that both suggested mechanisms; thickening and filter cake formation, could occur. External conditions such as dewatering rate and

temperature will in combination with the properties of the suspension, determine which mechanism would dominate. It was also found that the concentration gradient noticed in thin filter cakes of polydisperse size distribution could be caused by a size segregation effect rather than the proposed thickening mechanism, Figure 3.

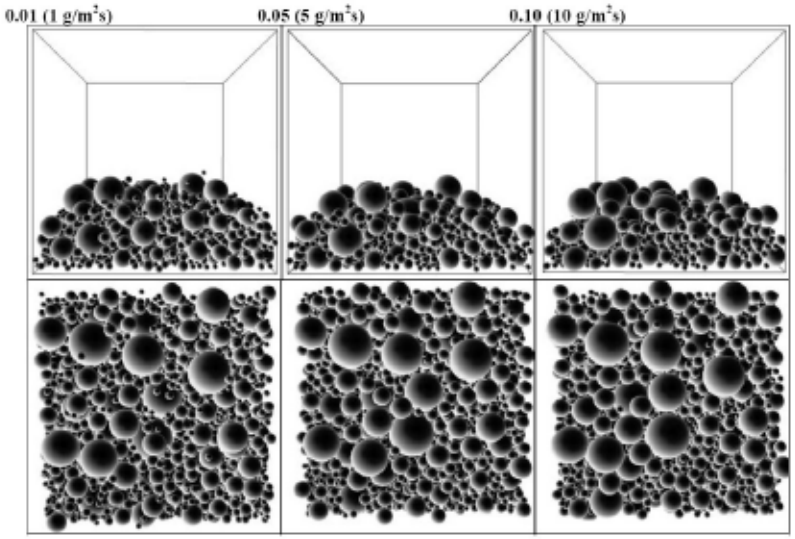

Figure 3. Size segregation in a filter cake composed of polydisperse particles at 3 different absorption rates. In this case, the filter cake is comprised of 1600 particles. Side view (above) and top view (below) [8].

A similar approach as presented in this work would be applicable for studies on, e.g., sedimentation rate depending on particle size and other system properties. It would also be possible to further the work on filtration processes and achieve a deepened understanding of prevalent mechanisms under various conditions and for various particle suspensions.

Colloidal interactions

DLVO-type colloidal interactions is considered to become significant if the size of particles is less than 10 µm. In many mineral processing applications such as comminution and particle separation processes, the particle size is often much larger and the consideration of these forces are not needed. However, the particle size distribution also needs to be taken into account. The small size fraction of a broad size distribution can fall under the colloidal size range, which in turn can influence the behaviour of the entire system [9]. One well known effect is a reduction of suspension viscosity as result of small particles acting as lubricants between larger particles.

In simulations there can be significant differences in particle behaviour, depending on if the colloidal forces are accounted for or not. For instance, Sand et al. [10] describes how the structure of consolidating calcium carbonate suspensions may be influenced by varying the colloidal interaction parameters. A typical effect is the formation of loose, rigid particle networks in systems dominated by van der Waals attraction. Conversely, repulsion dominated systems allow particles to arrange past each other and result in denser and less rigid particle networks. This behaviour is illustrated in Figure 4, where the consolidation of a fine-grade mineral suspension is simulated.

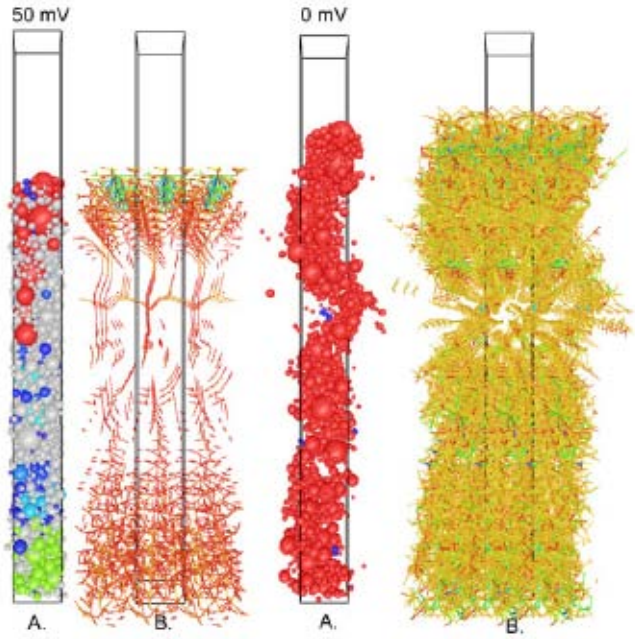

Figure 4. Dominance of electrostatic repulsion (50 mV particle surface potential) results in denser structure compared to van der Waals attraction (0 mV particle surface potential) dominated system. Figures A show particle colouring depending on cluster identity and figures B shows cluster network connections [8].

It was shown that even slight changes to DLVO-parameters can have influence on the solids concentration gradients and immobilisation times of suspensions and filter cakes. The accumulation of particles at the free surface of a mineral suspension during drying could to some extent be controlled by adjusting colloidal parameters [8].

The work illustrates the importance of understanding and controlling the colloidal properties of suspensions. A lack of understanding of these phenomena result in a number of process-related problems. Examples might include unwanted agglomeration, inhomogeneous distribution of additives, increased wear on equipment or abnormally high chemical consumption. These are factors which have not been extensively studied, except if necessitated by the constituents of the bulk.

Suspension rheology

The rheological behaviour of particle suspensions, or slurries, is complex. There can be variations in particle size, shape and concentration. The surface properties of particles and presence of additives will influence particle interactions and therefore also the macroscopic flow behaviour of the suspension. A suspension is typically non-Newtonian with rheological properties varying significantly with shear rate. The rheology of a suspension can be characterised empirically, but such investigations does not reveal the micro-level interparticle mechanisms that govern the behaviour of the particle-liquid system.

In this respect, particle dynamics simulation of slurry behaviour shows great promise. In the Stokesian dynamics approach presented in this work, suspension behaviour resulting from

hydrodynamic and colloidal interactions as well as adsorbed polymeric material can be investigated. The benefit of particle-level simulation, given proper model and parameter selection, is that most aspects related to the rheological properties of the slurry can come without preliminary assumptions regarding the slurry behaviour. This puts particle dynamics simulation in sharp contrast to continuum methods, where the rheological behaviour must be defined prior to simulation [2].

By Stokesian dynamics simulation of particles in shear and pressure-driven flows, it has been possible to report on rheology-related phenomena such as size segregation in polydisperse systems, lubrication effects by small particles in bimodal systems and dilatancy effects (structure jamming) in high solids concentration monodisperse systems [4,11]. The size segregation effect is illustrated in Figure 5.

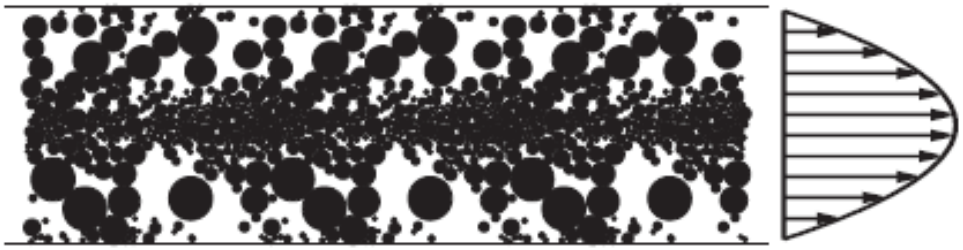

Figure 5. Example of size segregation in pressure-driven flow. Small particles preferentially migrate to the low shear rate region in the middle. Reproduced from [7].

4 CONCLUDING REMARKS

This paper presents a simulation approach for investigating 3D particle motion in high solids concentration, low particle Reynolds number colloidal suspensions. The analytical expressions for the hydrodynamic interactions were modified by fitting to results from finite element analysis. This allows for simulation of broad size distributions. The method is thus applicable for a wide range of particle systems and colloidal-level phenomena. These include e.g. microstructure development in time, particle motion mechanisms, and macroscopic flow properties as result of the micromechanical behaviour of particles. The current model does not back-couple particle movement to the liquid flow field. This assumption is however acceptable if particles are moving at a velocity comparable to that of the suspending medium.

4.1 Potential Applications of the Method in Mineral Processing

The modified Stokesian dynamics approach shows promise for simulation of high solids concentration fine particle solid-liquid systems, where particle interactions cannot be neglected. In addition, the validity of the Stokes law is assumed when describing the fluid's drag forces, thus limiting the method to non-turbulent fluid flow. Considering these constraints, several potential applications of the Stokesian Dynamics method can be identified within the field of mineral processing. Here fine and ultrafine solid particles often occur as intermediates and as final products from ore beneficiation and industrial minerals production. They can also occur as unintended by-products and are in such cases frequently regarded as a disturbing factor. Possibly with some method or model adjustments, the areas where Stokesian Dynamics is expected to be of beneficial use can be grouped as follows:

Slurry rheology

The flow properties of mineral slurries are known to affect the operation of a variety of wet mineral processing equipment and are thus of relevance for optimal design and process control. This is obvious for flow-based separation processes where the slurry flow properties determine the outcome, as is the case in particle size separation and gravity separation. Analysing various types of equipment and their ordinary operating conditions indicates possible applications for sluices and spiral separators, but also for wet shaking tables in which thin film flow is used to concentrate the heavier particles [12]. Other suitable separation processes are jigging, where stratification of a particle bed by particle density is achieved via applying a pulsating fluid flow to loosen the bed of particles, and upstream classification or upstream sorting respectively using a continuous fluid flow instead. In all these cases Stokesian Dynamics could provide a useful tool to identify parameter settings for adjusting slurry density and viscosity in order to improve efficiency of mineral separation and throughput.

Sedimentation and consolidation behaviour

Dewatering is an important ancillary process in most mineral processing operations [13]. Knowledge of the settling properties of fine particle mineral slurries is required when designing efficient dewatering systems for mineral concentrates and tailings by means of thickening and filtration processes. Particle simulations based on Stokesian Dynamics allow studying fine particle slurry dewatering for different equipment geometries and process parameters. Using appropriate non-hydrodynamic interaction models, even the influence of flocculants can be systematically investigated, in order to improve dewatering efficiency and increase unit capacity.

Particle agglomeration phenomena

As with decreasing particle size the surface and contact forces become more dominant compared to the body forces, adhesion of fine particles to other particles or walls has to be considered in several unit operations for mineral processing. Studying particle agglomeration occurring within fine particle processing is a therefore another area of application where Stokesian Dynamics based simulation can throw light upon the basic mechanisms of particle attachment and also detachment. This refers for instance to solids dispersion and mixing in the liquid phase, as well as the coating of grinding media and mill lining during comminution.

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